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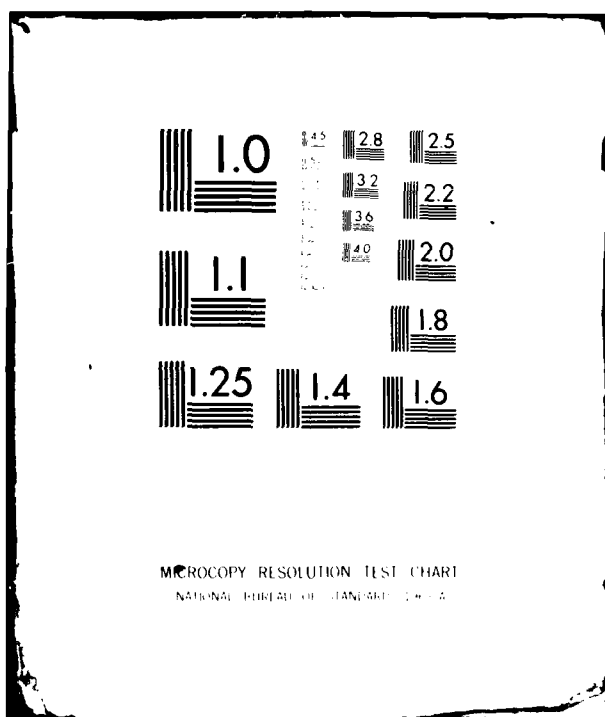
RELATIONSHIPS BETWEEN EXTINCTION AND MASS
CONTENT OF ATMOSPHERIC FOG AND MILITARY SMOKES, (U)

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INTRODUCTION:

A possible relation between the extinction (or
atmospheric visibility) and particle mass content of fogs and
aerosols has been a subject of interest to many scientists since
the beginning of the century.[26,1,6,13,25,3,17,19,10,2] If such a
relation exists, it would be of considerable practical interest
since then remote sensing extinction measurements would yield
information concerning the fog or aerosol mass content, and vice
versa, from mass content measurements (which are more easily done
at various meteorological stations) one could deduce the extinc-
tion properties of fogs and aerosols—at least at definite wave-
lengths.

All previous attempts to relate the extinction (or visi-
bility) to the mass content of fogs and aerosols were only par-
tially successful since all derived relations also turned out to
be functions of the particle size distribution, and thus the
derived relation varied from one case to another.

We will show that at definite wavelengths, which are
determined primarily by the size of the largest particles present
in a given polydispersion of fog or smoke aerosol particles, there
exists an unambiguous relation between the extinction and mass
content which is independent of the form of the particle size
distribution.

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EXTINCTION AND MASS CONTENT OF FOG AND SMOKE AEROSOL:

Consider a polydispersion of spherical aerosol particles characterized by a size distribution $n(r)$ and refractive index m. We want to derive relationships between the aerosol extinction coefficient σ_e and the aerosol mass content M given by

$$\sigma_e = \int \pi r^2 Q_e(m,x) n(r) dr \qquad (1)$$

$$M = \rho \int \frac{4}{3} \pi r^3 n(r) dr \qquad (2)$$

where $Q_e(m,x)$ is the Mie efficiency factor for extinction for a particle with refractive index m and size parameter $x = 2\pi r/\lambda$ at the radiation wavelength λ and ρ is the aerosol density.

It can be seen from the integrals in equations (1) and (2) that for the extinction coefficient σ_e and mass content M to be uniquely related for arbitrary particle size distributions $n(r)$, we must be able to approximate the Mie extinction efficiency factor $Q_e(m,x)$ by a linear function of particle radius $Q_e \backsim r$ [or equivalently, let us say $Q_e(x) = cx$ as suggested by Chýlek].[4,5] Then the right-hand side of equation (1) contains the integral $\int r^3 n(r) dr$ and combining equations (1) and (2) yields

$$\sigma_e = \frac{3\pi c}{2\lambda \rho} M . \qquad (3)$$

Unfortunately, the task of finding a wavelength for which the $Q_e = cx$ approximation is a good one is formidable because in general the extinction efficiency is a rather complicated function of particle size, refractive index, and wavelength. Nonetheless, examination of the form of the Mie efficiency function $Q_e(x)$ for fog droplets, phosphoric acid droplets, HC, and carbon smoke particles for wavelengths ranging from the visible through the middle infrared reveals that at particular wavelengths the $Q_e(x) = cx$ approximation is acceptable.

An example of such a wavelength for fog droplets is shown in figure 1 where the Mie extinction efficiency function at $\lambda = 11\mu m$ is well approximated by $Q_e = cx$, up to a maximum value $x_m \cong 8$

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(corresponding to a maximum radius $r_m \cong 14 \mu m$). Therefore, under the constraint that fog droplets have radii not exceeding $r_m = 14 \mu m$ (which is not unrealistic for radiation and most advection fogs), we expect a linear relation between extinction at $\lambda = 11 \mu m$ and liquid water content of fog according to equation (3).
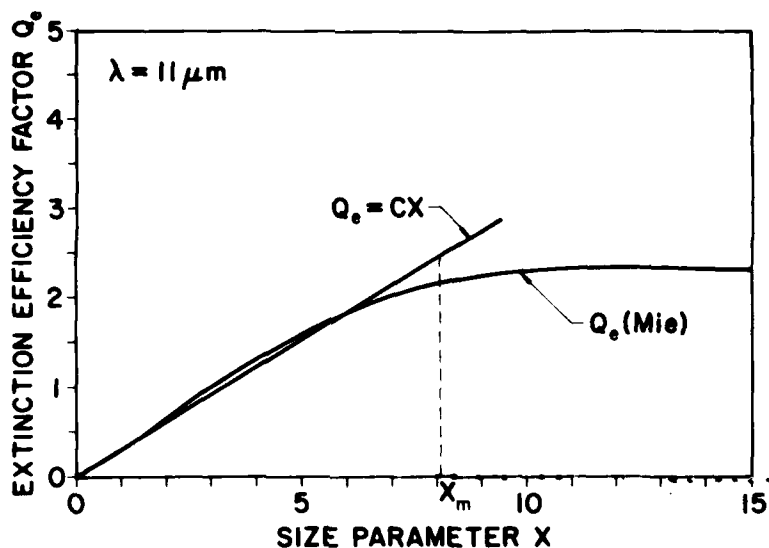

Figure 1. The efficiency factor for extinction Q_e for water droplets versus size parameter x at a wavelength λ = 11μm (index of refraction m = 1.153 - 0.0968i). The efficiency factor can be approximated by a straight line Q_e = cx providing $x \leq x_m$. The approximation overestimates the exact value of Q_e for some some size parameters, but underestimates it for others (still with $x \leq x_m$). These two errors tend to cancel, leading to the extinction coefficient being linearly related to the liquid water content of fogs according to equation (3).

    We should point out that this linear approximation of the Mie extinction efficiency is distinctly different than a Rayleigh approximation, which requires the particle size (with respect to wavelength) and phase shift be small ($|m - 1|$ x << 1). Clearly, this Rayleigh condition is not satisfied for size parameters as large as x = 8 as in figure 1.

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In the next sections we apply this approximation procedure and the resulting extinction-mass content prediction (3) to polydispersions of particles having widely different optical properties: atmospheric fog, phosphoric acid, HC, and carbon smokes. The success achieved with relation (3) attests to its simplifying impact on the gamut of radiative transfer problems in which atmospheric fog or aerosols play a role. Thus, our relation (3) connecting particulate extinction to particulate mass content should prove useful in such diverse research areas as earth climate, remote sensing, and electro-optical communications.

APPLICATION TO ATMOSPHERIC FOG:

Before we can have confidence in applying the linear relation (3) between extinction and liquid water content of fog we should test its validity with existing measurements that are available. Since simultaneous infrared extinction and liquid water content measurements in fog do not yet exist, we calculated the volume extinction coefficient σ_e using Mie theory and the liquid water content M for 341 measured size distributions of fog droplets reported in the literature.[9,10,22,13,14,21] The fog measurements were chosen to represent a wide range of meteorological conditions ranging from maritime and continental advection fogs[13,14,9] to inland radiation fogs.[9,10,22,21]

The results of these calculations together with the derived approximation equation (3) at $\lambda = 11\mu m$ are shown in figure 2. Points showing the results of the Mie calculations are always within a factor 2 of the straight line showing the equation (3) approximation ($\sigma_e = 128W$) even though the extinction coefficient and liquid water content ranges over almost four orders of magnitude with the considered size distributions. In view of the fact that the $\sigma_e = 128W$ relation is not a result of any fit to experimental points, but rather an absolute calculation (by absolute we mean that no free fitting parameter is involved) using equation (3), we consider the agreement to be acceptable.

On the other hand, we do not expect a unique extinction-liquid content relationship at all wavelengths. To demonstrate this point, we present Mie calculations for the same 341 droplet distributions at $\lambda = 4\mu m$ in figure 3. For a given liquid water content, the extinction coefficient varies by more than an order of magnitude as a function of the size distribution.
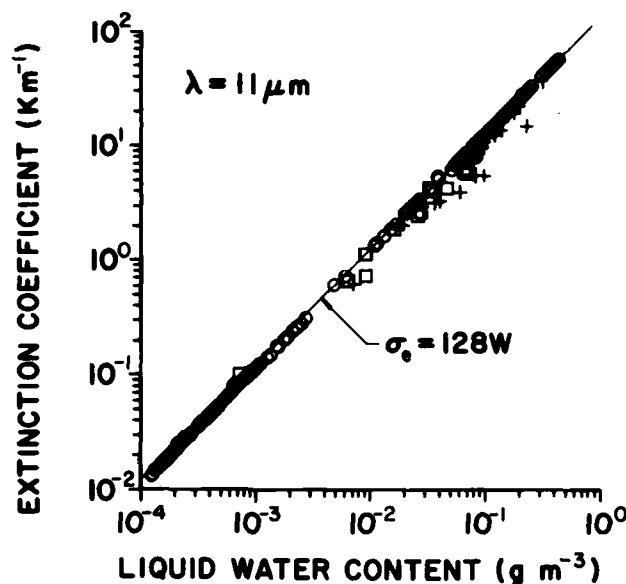
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Figure 2. Variation of particulate extinction coefficient (at λ = 11μm) with liquid water content W. The points were determined from 341 size distribution measurements of atmospheric fog and haze made at different geographic locales and under a variety of meteorological conditions [open circles denote measurements of Pinnick,[21] plus symbols of Garland and Roach[9,10,22], square symbols of Kumai,[13] and solid circle symbols of Kunkel[14]]; the straight line labeled σ_e = 128W was determined from equation (3).

APPLICATION TO PHOSPHORIC ACID AND RP SMOKE:

Phosphoric acid smoke differs from atmospheric fog in two important respects. First, smoke particles are much smaller than fog droplets; and second, they are generally strongly absorbing at infrared wavelengths. These two characteristics result in Mie efficiency factors Q_e being well approximated by Q_e = cx for most infrared wavelengths, and consequently, we expect the extinction-mass relation (3) should be applicable throughout the infrared.
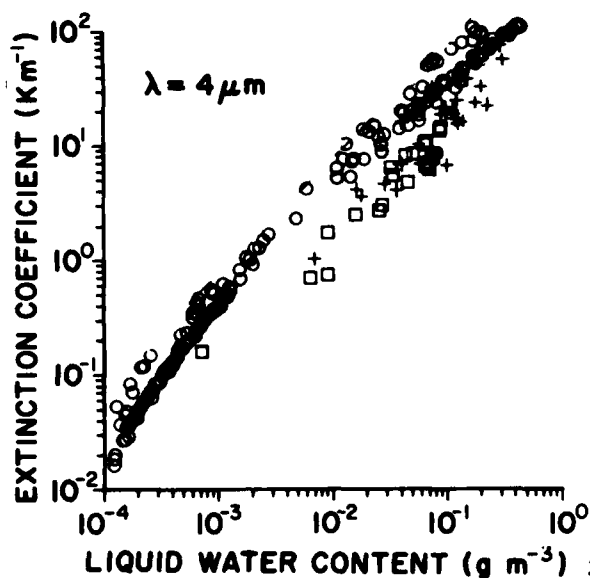
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Figure 3. Variation of particulate extinction coefficient (at λ =
4 μm) with liquid water content for the 341 size distri-
butions of fog and haze considered in figure 2.

To test this contention we have compared the ratio of the
extinction coefficient to mass content σ_e/M predicted according to
our size-distribution-independent relation (3) to values of σ_e/M
determined from the transmission and mass content measurements of
Milham.[1 5]* The results, which are seen in figures 4 and 5, show

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*The extinction coefficients σ_e were derived from transmission
measurements for which forward scattering corrections[7] and
multiple scattering corrections should be considered. Forward
scattering corrections arise from singly scattered photons that
enter the detector along with the unscattered (direct) radiation
due to the finite angular aperture of the detector. Similarly,
multiple scattering corrections arise from signal contributed by
multiply-scattered photons. Both these effects cause increased
detector signal and hence result in a smaller inferred extinction
coefficient if they are not considered. We estimate the forward
scatter corrections for the experimental setup used by Milham[1 5] to
be not more than 3 percent and have neglected them; however, no
attempt was made to make quantitative estimates of multiple
scatter corrections.

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agreement of predicted and measured σ_e/M values to within 30 percent throughout the 3mm to 5µm and 8µm to 12µm atmospheric window spectral regions suggesting the validity of relation (3) for phosphoric acid smoke. The extinction-mass measurements in figures 4 and 5 can only suggest (and not prove) the validity of relation (3), as the measurements are for only one particular size distribution of phosphoric acid particles.
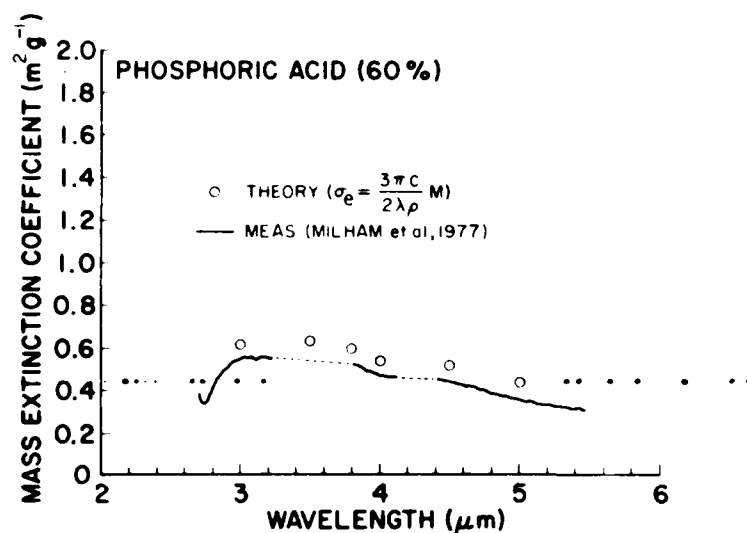

Figure 4. Values of the ratio of aerosol extinction coefficient to aerosol mass content (σ_e/M in m^2g^{-1}) predicted according to the size-distribution-independent linear relation (3) (open circles), and measured by Milham[15] (smooth curve) for 60 percent phosphoric acid aerosol. Good agreement between prediction and measurement is obtained throughout the 3µm $\leq \lambda \leq$ 5µm spectral region.

In contrast to the phosphoric acid results, comparison of the relation (3) to Milham's[15] measurements on RP smoke (figure 6) show relatively poor agreement in the 8µm to 12µm spectral region. The reason for the poor agreement is that the burning of RP/WP smokes apparently results in production of an unknown chemical species[16] whose refractive indexes cannot be approximated by those of phosphoric acid [which was assumed in determination of $c(\lambda)$ in relation (3)].
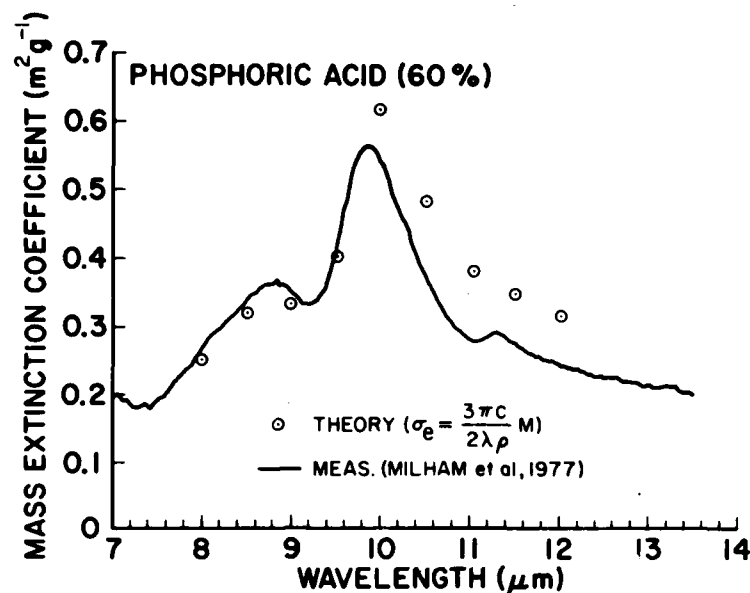
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Figure 5. Same as figure 4 except for the $8\,\mu m \leq \lambda \leq 12\,\mu m$ spectral region.

APPLICATION TO HC SMOKE:

Like phosphoric acid and RP smoke particles, HC smoke particles are much smaller than fog droplets. However, they are much weaker absorbers than these other smokes at most infrared wavelengths, and as a result the $Q_e = cx$ approximation is less accurate.

This inaccuracy is reflected in the comparison of σ_e/M values predicted according to equation (3) with measurements of Milham[15] in figures 7 and 8. For some wavelengths in the $3\,\mu m$ to $5\,\mu m$ and $8\,\mu m$ to $12\,\mu m$ spectral regions the disagreement between prediction and measurement is as much as a factor 2.5. Thus, although we expect relation (3) to be successful throughout the infrared for strongly absorbing phosphoric acid smoke, for weakly absorbing HC smoke our relation (3) should only be applied for wavelengths around $\lambda = 4\,\mu m$ and $\lambda = 11\,\mu m$.

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RP SMOKE (65%)

THEORY $(\sigma_e = \frac{3\pi C}{2\lambda\rho} M)$

MEAS.(MILHAM,1976;DISSEMINATION BY BURNING)

——— ML = 1.07 g m^{-2}
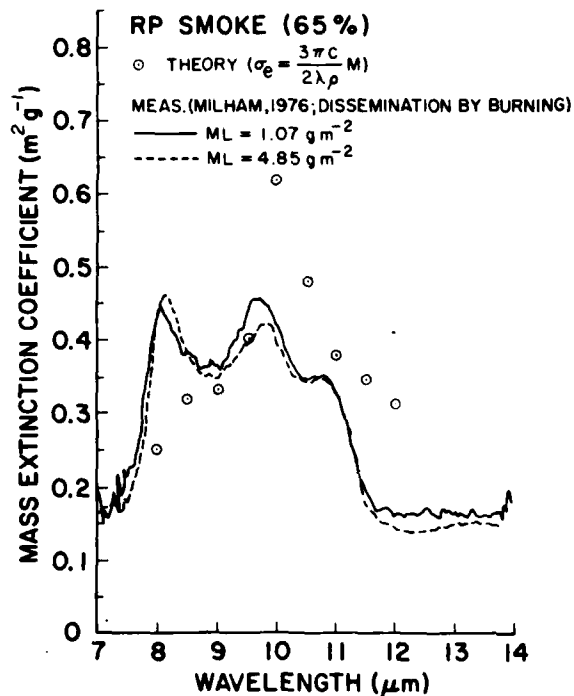
------- ML = 4.85 g m^{-2}

Figure 6. Same as figure 5 except for RP smoke rather than phos-
phoric acid. In this case, measurements are shown for
two different aerosol mass loadings (M is the aerosol
mass content and L the transmission chamber path
length).

APPLICATION TO CARBON SMOKE:

Caution must be exercised in applying the aerosol extinc-
tion coefficient-mass content relation (3) to carbon smoke because
in general the particles are irregular in shape and the derivation
of equation (3) requires the assumption of spherical homogeneous
particles. We know from extinction and scattering measurements on
slightly irregular particles by Greenberg,[11] Zerull,[27] and
Pinnick,[20] that providing their equivalent size parameters are
less than x = 3, they scatter like spheres. If carbon smoke
particles were only slightly irregular and if they had size param-
eters less than x = 3, we might expect that our neglect of irreg-
ular particle effects in the application of equation (3) to these

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particles might be of little consequence. However, some carbon smoke particles, for example those measured by Roessler and Faxvog,[23] consist of long extended chains of small (∿0.05μm diameter) spheres (figure 9). For these particles, we are probably not justified in approximating their extinction efficiency by some equivalent sphere extinction efficiency [as is done in determination of the parameter c in equation (3)].

Keeping in mind reservations concerning particle shape effects, we have proceeded to compare the extinction coefficient-mass relation (3) to transmission measurements made through carbon smokes from several different sources. We chose measurements reported by Hurley and Bailey[12] on a hand-fired Lancashire boiler operating at full load, by Conner and Hodkinson[6] on an experimental smoke stack, by Roessler and Faxvog[23] on acetylene smoke, and from the Handbook of the Society of Automotive Engineers (SAE),[24] for diesel engine smoke emissions.
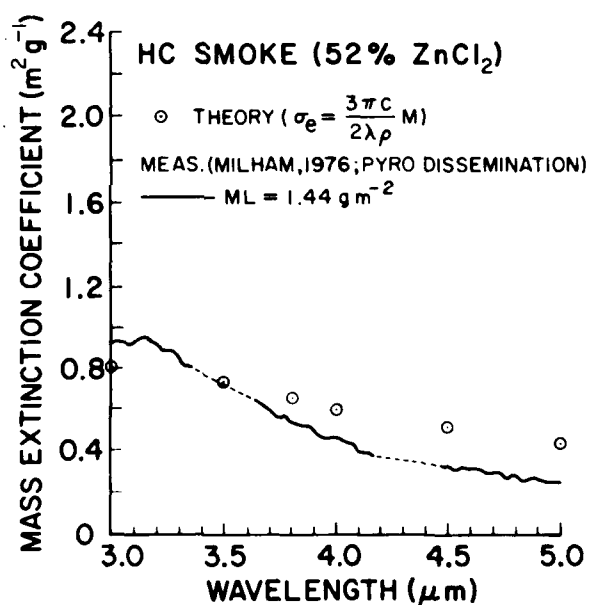
HC SMOKE (52% $ZnCl_2$)

⊙ THEORY ($\sigma_e = \frac{3\pi c}{2\lambda\rho}$ M)

MEAS.(MILHAM,1976;PYRO DISSEMINATION)

—— ML = 1.44 g m^{-2}

Figure 7. Values of the ratio of aerosol extinction coefficient to aerosol mass content (σ_e/M in m^{2}g^{-1}) predicted according to the size-distribution-independent relation (3) (open circles), and measured by Milham[15] (smooth curve) for HC smoke.
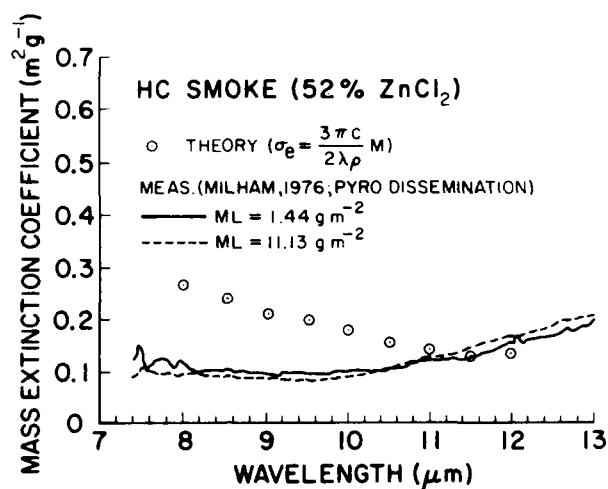
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Figure 8. Same as figure 7 except for the $8\,\mu m \leq \lambda \leq 12\,\mu m$ spectral region. In this case, measurements are shown for two different aerosol mass loadings (M is the aerosol mass content and L the transmission chamber path length).
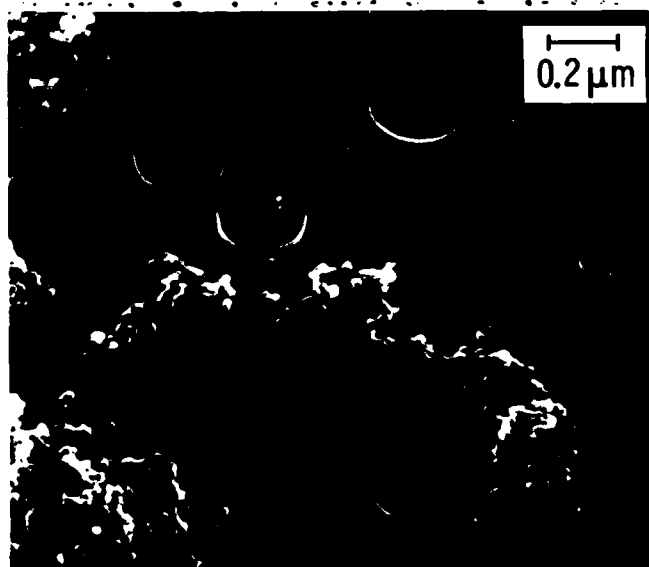

Figure 9. Scanning electron microscope micrograph showing carbon smoke particles collected onto a Nuclepore filter (from Roessler and Faxvog).[23]

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The results of the comparison are shown in table 1 where the values of σ_e/M predicted on the basis of equation (3) are generally within 20 percent of those determined from the measurements on various types of carbon smokes. Thus, even for quite nonspherical smoke particles, the relation (3) correctly predicts a relation between extinction and mass content, at least for visible wavelengths.

CONCLUSIONS:

We have shown that a linear size-distribution-independent relation should exist between the volume extinction coefficient and particulate mass content of atmospheric fog, phosphoric acid smoke, HC smoke, and carbonaceous smokes--but only at particular wavelengths. The suitable wavelengths are $\lambda \cong 11\,\mu m$ for fog, $\lambda \cong 3\,\mu m$ to $5\,\mu m$, $8\,\mu m$ to $12\,\mu m$ for phosphoric acid, $\lambda \cong 4\,\mu m$, $11\,\mu m$ for HC, and $\lambda \cong 0.55\,\mu m$ for carbon smoke. These relations can be used to uniquely connect the transmission across a path in a fog or smoke cloud to the path-integrated particulate mass content, providing forward scattering and multiple scattering corrections can be neglected.

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TABLE 1

Comparison of measured and predicted values of the ratio of
aerosol volume extinction coefficient to mass concentration σ_e/M
for carbonaceous smoke particles. The measured values of σ_e were
determined from visible transmissometer measurements under the
assumption that forwardscatter and multiple scatter corrections to
the inferred extinction coefficient are negligible. The measured
values of smoke mass concentration M were determined from filter
sampling. Also given are values of the smoke path length L and
the transmissometer wavelength λ. The predicted values of σ_e/M
were obtained from the size-distribution-independent relation (3)
under the assumption of a particle density $\rho = 2$ g cm^{-3}.

Reference	Mass Concentration $M(g\ m^{-3})$	Path Length $L(m)$	Wavelength $\lambda(\mu m)$	σ_e/M (m^2g^{-1})	Predicted [(from equation (3)] $\sigma_e/M = 3\pi c/2\lambda\rho$ (m^2g^{-1})
Black Smoke (coal stoker) Hurley and Bailey (1958)	0.29	0.305	0.55	8.04	9.5
	1.05	0.305	0.55	8.38	
	1.40	0.305	0.55	9.0	
Black Smoke (oil fired smoke stack) Conner and Hodkinson (1967)	0.16	0.10	0.56	8.78	9.3
	0.55	0.10	0.56	9.61	
	1.0	0.10	0.56	9.52	
Acetylene smoke Roessler and Faxvog (1979)	0.0155	1.835	0.514	9.93	10.2
	0.0527	0.10	0.514	9.96	
	0.120	1.835	0.514	9.68	
	0.165	0.10	0.514	9.60	
	0.321	0.10	0.514	10.1	
Diesel Engine Emission SAE handbook (1977)	1.0	0.10	0.55	6.79	9.5

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